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Emissions from Cofiring Surrogate Hazardous
Waste in a Commercial Packaged Boiler

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Emissions From Cofiring Surrogate Hazardous
Waste in a Commercial Packaged Boiler

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Steady state and transient operating conditions were studied when cofiring surrogate liquid hazardous waste in a firetube packaged boiler. The destruction and removal efficiencies (DREs) for the principal organic hazardous constituents (POHCs) were determined under varying combustion conditions. Results indicate that aromatic chlorohydrocarbons are the most difficult to destroy. A DRE less than 99.99% was experienced for transient operating conditions in load, stoichiometry, and waste feed rate. Steady operation above 10% excess air ensures DREs greater than 99.99%. Major products of incomplete combustion (PICs) measured were methylene chloride and chloroform.

INTRODUCTION

Cofiring or co-combustion of hazardous compounds with conventional fuels in industrial boilers is widely practiced in the U.S. Since this use of waste with a high heating value as a fuel represents an attractive energy recovery option, it has historically been exempted from Resource Conservation and Recovery Act (RCRA) controls. Environmentally, thermal destruction of hazardous waste is preferable to landfilling of these compounds if destruction is effective, but a sufficient data base indicating successful destruction is needed. In addition, hazardous materials can be destroyed on-site thus eliminating the problems of storage and transportation. However, these hazardous waste compounds should be destroyed in the boiler with the same effectiveness as required of an incinerator. Incinerator rules are designed to limit atmospheric emissions of the principal organic hazardous constituents (POHCs) and require that these systems have a destruction and removal efficiency (DRE) of $\geq 99.99\%$. The DRE is defined by:

$$DRE = (M_f - M_s) / M_f \times 100$$

where

M_f = mass flow rate of a POHC in the waste, and
 M_s = stack mass emission of a POHC.

With the growing concern over possible environmental impact, EPA is developing regulations to cover cofiring of hazardous waste materials. There remains a need to characterize DRE during cofiring under nonstandard and transient conditions. Such conditions could be the result of facility start-up, soot blowing, load change, hardware malfunction, overcharging, etc., and could lead to failure modes of operation where DRE is less than 99.99%.

This paper is a result of a study sponsored by EPA's Combustion Research Branch in the Air and Energy Engineering Research Laboratory (AEERL) to examine the effects of boiler cofiring of surrogate waste on emissions, including those under nonstandard operating conditions.

EXPERIMENTAL FACILITY AND METHODS

The primary component of the test facility (see Fig. 1) was a North American Atlas brand, 0.8 MW (2.7 million Btu/hr), three-pass firetube package boiler capable of firing either fuel oil mixtures and/or natural gas. The boiler support systems were designed for independent control of combustion air, fuel(s), and air atomization. The boiler load was passed to a shell and tube heat exchanger, which subsequently transferred the load to an evaporative cooling tower loop.

Another major component of the test facility was the surrogate waste feed system. The waste mixture was contained in a steel drum, which was located on an electronic weigh scale for measurement of average waste consumption rates. From the drum the waste mixture was pumped into the boiler through a volume flowmeter and a manual flowmetering valve.

Stack concentrations of O_2 , CO_2 , CO , and NO were measured by a Continuous Emissions Monitoring (CEM) System.

A variation of the Volatile Organic Sampling Train (VOST) procedure, called mini-VOST [1], was used to characterize the stack emissions of chlorinated hydrocarbons. Ten liter samples, in duplicate, were taken for the various data points. Tenax tubes, used to absorb chlorinated hydrocarbons, were analyzed within 24 hr of sampling.

APPROACH

The primary boiler fuel used was natural gas. Initially, boiler gaseous emissions were characterized under baseline (no POHC) cofiring steady state conditions. Carbon tetrachloride (CCl_4), monochlorobenzene (MCB), and perchloroethylene (PCE) were chosen to be the POHCs used in this study. The surrogate hazardous waste mixture used was 94% distillate fuel oil and 2% by weight each of CCl_4 , MCB, and PCE. The composition and the amount of waste mixture cofired with natural gas were based on the range of heating values of the individual species and on composition of hazardous waste mixtures encountered in practice.

Under steady state cofiring conditions, the nominal boiler operating parameters of stoichiometric ratio (SR) and firing rate were chosen to be 1.10 SR and 0.64 MW (2.2 million Btu/hr), respectively. The percentage of boiler heat input contributed by the waste mixture determined the fuel heating value. The nominal value for this parameter was chosen to be 25%, corresponding to a fuel higher heating value (HHV) of 51.8 MJ/kg (22.3 kBTu/lb). Subsequently, parametric tests were carried out to study the effect of boiler SR, firing rate, fuel heating value, and degree of waste mixture atomization on the DRE under steady state nonstandard boiler operation. Further tests were carried out to characterize the effects of transient boiler operating conditions on the DRE. The transient boiler conditions of load, SR, and waste flow rate were chosen for experimental investigation because these transient conditions occur in commercial practice.

Products of incomplete combustion (PICs) could be just as harmful as, or even more harmful than POHCs; therefore, PICs were also screened during the analysis of the mini-VOST samples.

RESULTS

DRE Versus Stoichiometric Ratio

The effect of SR on DRE is depicted in Fig. 2. Operation at relatively low SRs could lower DREs due to oxygen deficiency, as shown by the data points for 80% firing rate and the lower HHV. Operation at relatively high SRs at constant load could lower the temperature of the boiler and decrease the combustion gas residence time, resulting in a decrease of the destruction of POHCs and lower DREs. Based on these counter effects, a maximum in the DRE versus SR curve would be expected. A slight indication of this trend is exhibited by the 80% firing rate and the 51.8 MJ/kg (22.3 kBTu/lb) HHV data.

DRE Versus Firing Rate

An increase in firing rate corresponds to a decrease in residence time of the combustion gases in the boiler. This could, potentially, lead to a decrease in the DRE for POHCs. The results for this effect are plotted in Fig. 3. The effect is indicated for three of the four tested combinations of SR and HHV. The exception is 1.1 SR and 51.8 MJ/kg (22.3 kBTu/hr), respectively, where no change is noted with firing rate. With 10% excess air and a higher heating value of 51.8 MJ/kg (22.3 kBTu/hr), perhaps the POHC destruction reaction kinetics are fast enough to offset the residence time decrease. Only the 1.0 SR and lower HHV tests indicated a serious failure mode potential.

DRE Versus Fuel Higher Heating Value (HHV)

The effect of decreasing the fuel HHV (by increasing the amount of low heating value waste content in the fuel) at constant SR and firing rate would be a decrease in the DRE via a decrease in the temperature of combustion gases. These trends are seen in Fig. 4. The DRE decrease, with a decrease in fuel heating value, is more dramatic with a SR of 1.0 perhaps due to additional effects of oxygen paucity.

DRE Versus Degree of Waste Mixture Atomization

The degree of atomization affects the combustion efficiency by decreasing/increasing the surface area of the fuel available for combustion. Poor atomization would lead to a decrease in surface area available for combustion and would be expected to lower the POHC DREs. Two atomization states were established for the waste mixture by creating two atomizing air flows to the atomizing nozzle. One of these states was "good" atomization and the other "poor," both of which were established visually. The atomization states were varied at nominal furnace conditions, and the results in Fig. 5 show the lowering of DRE with deterioration in atomization quality of the waste mixture.

Transient Ramping of Firing Rate

Maintaining the SR at 1.1 and the waste flow rate constant under relatively good atomization conditions, the firing rate was ramped up from a baseline of 70% to about 90% and then ramped back down to 70%. The processes of ramping up and down were each completed at a constant rate in 15 min and, during each ramping condition, mini-VOST samples were taken continuously over the entire period of the ramp. The test results are plotted in Fig. 6. On ramping up, the residence time of the combustion gases decreases and the total POHC DRE drops slightly; on ramping down the reverse happens and DRE increases slightly. The effect, if any, between the two baseline DREs at the beginning of ramping up and the end of ramping down is interesting and needs to be substantiated with more data.

The test results using a more rapid rate, 10 rather than 15 min, are also plotted in Fig. 6. The difference in the two baselines (beginning and end of transient conditions) seems to be greater, suggesting that perhaps the effects of lower residence times during ramp up continued into the ramp down conditions, with a greater effect during a relatively rapid transient phase. Thus, the time slope of the ramp up/ramp down condition seems to be related to DRE, but this needs to be substantiated by further study.

Transient Fluctuations in SR

At nominal conditions of firing rate and waste HHV, the boiler SR was varied periodically between 1.1 and 1.0. This was done for two wastes, corresponding to fuel mixture HHVs of 51.8 and 48.8 MJ/kg (22.3 and 21.0 kBtu/lb). The test results are plotted in Fig. 7. Also plotted in Fig. 7, for comparison, are the relevant baseline results. The decrease in DRE during transient conditions could be attributed to the sensitivity of DRE to SR. Also, the SR may have gone down even below 1.0 during transient conditions, thus giving a DRE even lower than that at steady state and a SR of 1.0. No trend seemed apparent on changing the waste heating value from 51.8 to 48.8 MJ/kg (22.3 to 21.0 kBtu/lb). It is worth noting that transient fluctuations in SR could easily lower DRE values to below 99.99%, as indicated in Fig. 7.

Transient Fluctuations in Waste Flow Rate

Starting with nominal steady state operation of the boiler, the waste flow rate was varied periodically such that the waste heat input was between 30 and 25%. The test results are shown in Fig. 8. Also plotted in Fig. 8, for comparison, are the relevant baseline results. The variation in waste flow rate may have caused a combination of poor atomization (due to pulsating flow to the nozzle) and oxygen deficiency per mole of POHC to decrease the DRE. It is worth noting that the POHC DRE went down below 99.99% for the tests performed.

Individual POHC DRE Comparisons

To explore the effect of molecular structure of POHC on DRE, DREs for the individual POHC are cross-plotted in Figs. 9-11. The 45 degree line drawn gives an indication of relative ease/difficulty of destruction. These plots suggest that the PCE was destroyed more easily than CCl_4 , while CCl_4 was destroyed more easily than MCB. Thus MCB seemed to be the hardest to destroy among the POHCs tested.

Performance Indicators (DRE versus NO, CO)

Carbon monoxide (CO) and nitric oxide (NO) are emitted from all combustion systems in varying amounts. As CO is an indicator of the degree of completion of combustion, and NO is an indicator of temperature/SR conditions in the furnace, these measures have been considered as possible indicators of boiler performance. Several studies have been conducted on this aspect [2,3], and it has been concluded that the relationships between DRE and these parameters are, in all likelihood, facility specific.

In this study, correlations between DRE and CO/NO emissions were attempted. The results are plotted in Figs. 12 and 13. The plots indicate that DRE increases with increasing NO emissions, perhaps due to increasing temperature, and DRE decreases with increasing CO emissions due to worsening combustion conditions.

PIC Emissions

Table I summarizes the chlorinated PIC and POHC emissions for the various data points analyzed by mini-VOST during this study. It can be seen that the PIC concentration increases with an increase in POHC concentrations. For the range of parameters tested, the PIC/POHC ratio varied between 0.03 and 8.87, giving mean and median values of 1.14 and 0.67, respectively, thus indicating that PIC concentrations were of the same order of magnitude as the POHC concentrations. As chlorinated PICs could be just as hazardous as POHCs and their emissions are of the same order of magnitude, they deserve as much attention as POHCs from an environmental pollution standpoint.

Potential Failure Modes

Table II summarizes all the experimental conditions at which the POHC DRE was found to be very near or below 99.99%.

CONCLUSIONS AND RECOMMENDATIONS

Since this study was of a preliminary nature, the conclusions and recommendations are not heavily supported by data quantity. They are offered as a potential guideline for further study. Analysis of the results of the various tests performed in this study yielded the following conclusions and recommendations:

- (a) As load and waste heat input were increased, and SR was decreased, the POHC DRE was observed to decrease within the range of parameters tested.
- (b) SR, or oxygen available per mole of POHC, was a parameter affecting the POHC DRE under steady state, good atomization conditions. At a SR below 1.1, the POHC DRE can potentially be below 99.99%. The effect is especially noticeable with MCB.
- (c) Under the conditions investigated, MCB was the hardest to destroy among the three POHCs tested. This may suggest that aromatic chlorinated hydrocarbons could potentially pose emission problems in cofiring. However, this observation needs to be further corroborated by careful experimentation.
- (d) Based on the two conditions tested, the degree of atomization seemed to affect the POHC DRE. Even at excess air levels of 10%, poor atomization caused the POHC DREs to be of the order of 99.9%. Thus, it would seem useful to study the effect of atomization on POHC DRE in more detail.
- (e) There seemed to be a direct relationship between POHC DRE and NO emissions, and an inverse relationship between POHC DRE and CO emissions. However, much more data would be needed to determine if there is a strong correlation between these parameters.

(f) For the conditions tested, transient firing rates never resulted in POHC DREs being less than 99.99%. There was no conclusive evidence that upward or downward ramping yielded significantly different DREs. Furthermore, the ramping rate did not seem to significantly affect the DRE. However, more experimentation is recommended to corroborate this conclusion.

(g) For both test points with transient SRs the POHC DREs were below 99.99%. Thus, transient SRs seem to have a major influence on DRE.

(h) At the one condition tested with a transient waste flow rate, the POHC DRE was found to be below 99.99%, perhaps implying that transience in flow rates is also a critical parameter.

(i) Summarizing for the treatment conditions tested, waste flow rate and SR seemed to have a greater impact on DRE than did a transient firing rate. More data could corroborate this.

ACKNOWLEDGMENT

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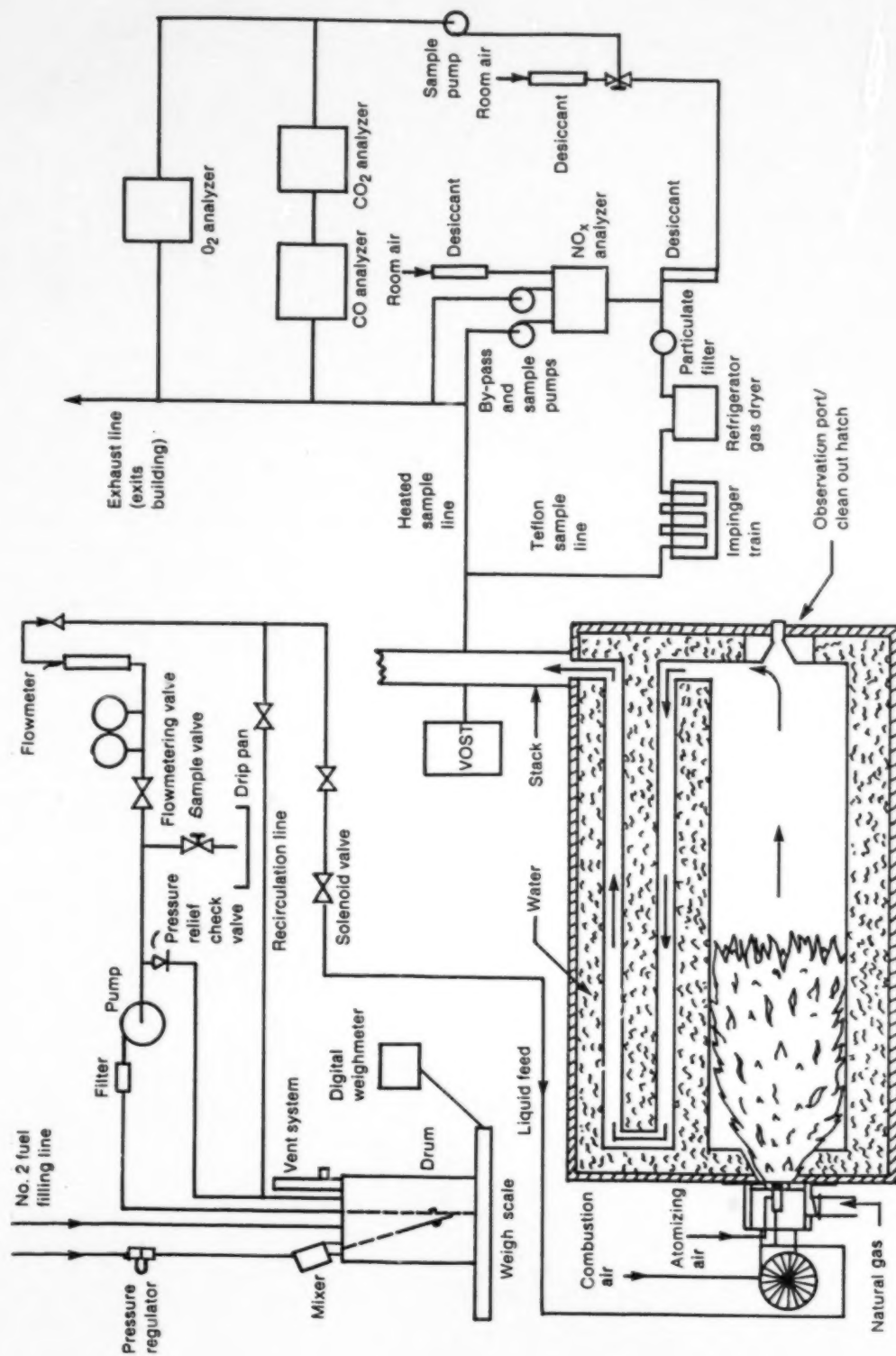


FIG. 1 EXPERIMENTAL FACILITY

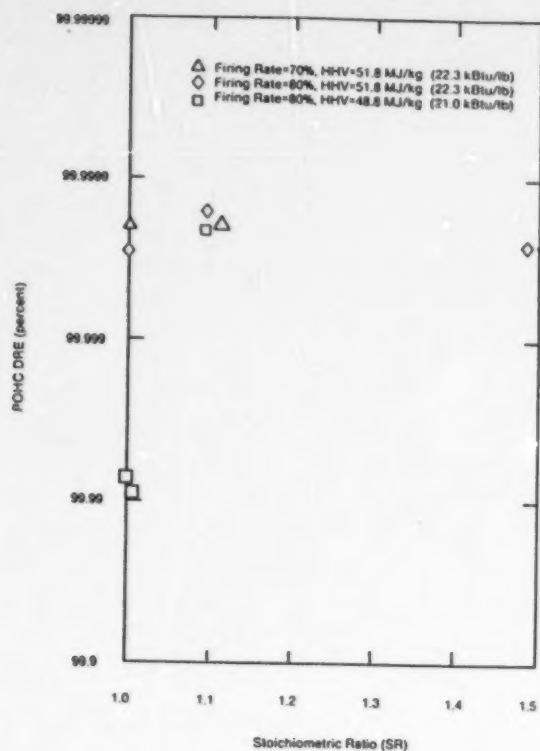


FIG. 2 POHC DRE VERSUS STOICHIOMETRIC RATIO (SR)

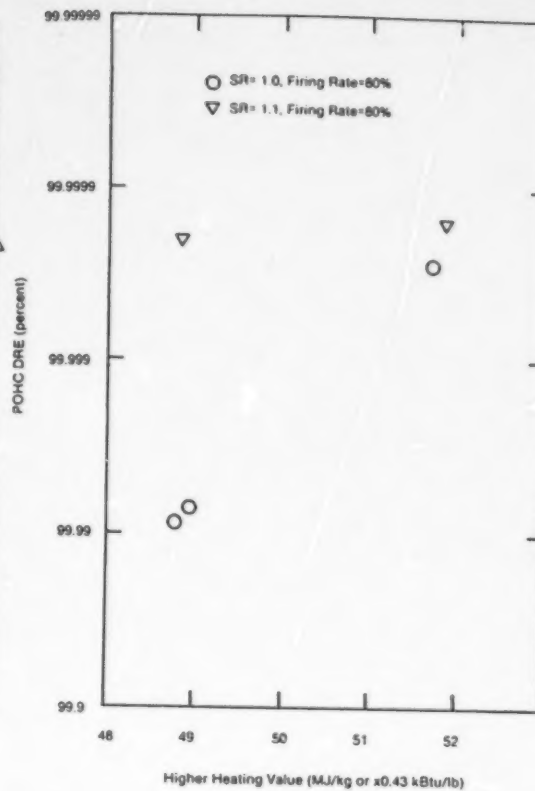


FIG. 4 POHC DRE VERSUS FUEL HEATING VALUE

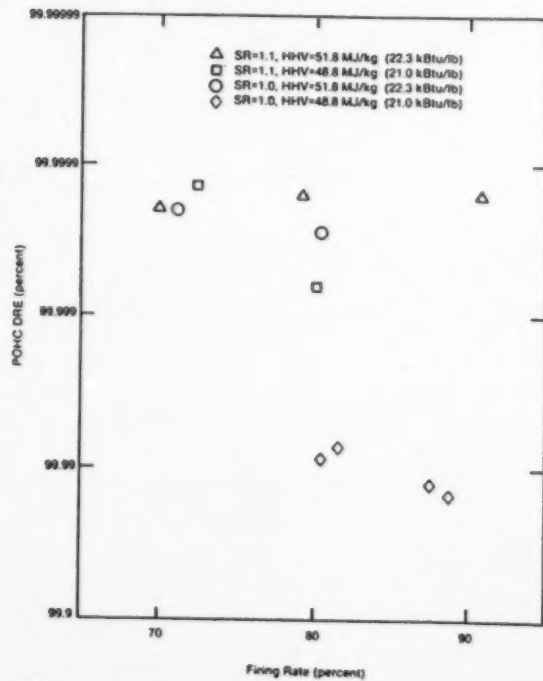


FIG. 3 POHC DRE VERSUS FIRING RATE

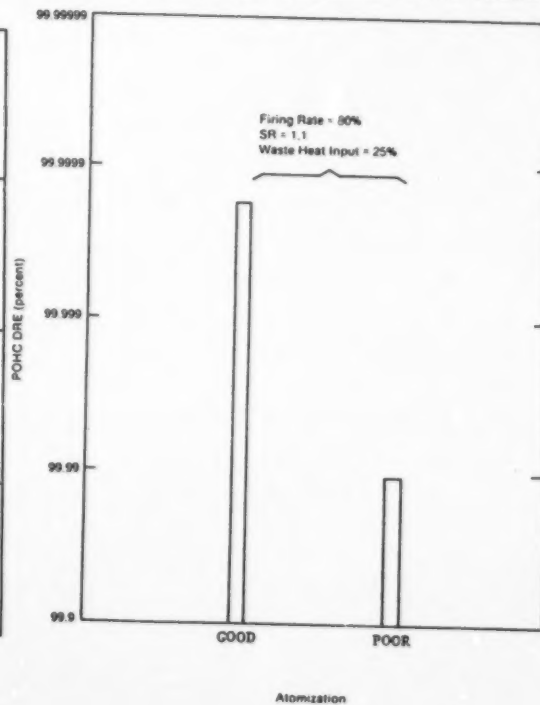


FIG. 5 POHC DRE—GOOD ATOMIZATION VERSUS POOR ATOMIZATION

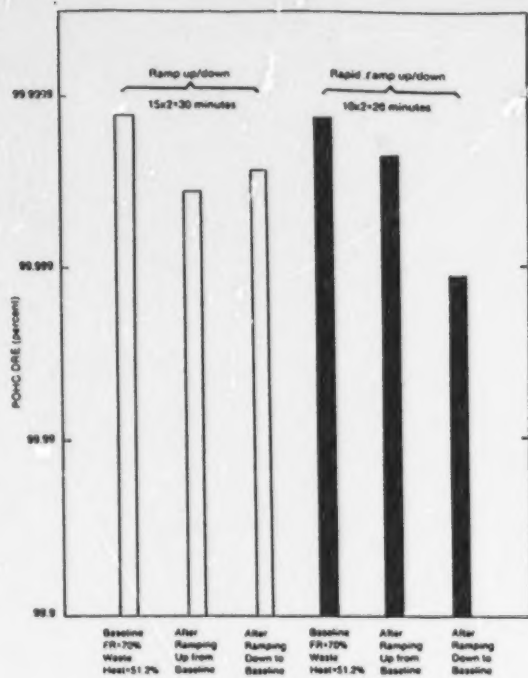


FIG. 6 POHC DRE VERSUS RAMPING OF FIRING RATE

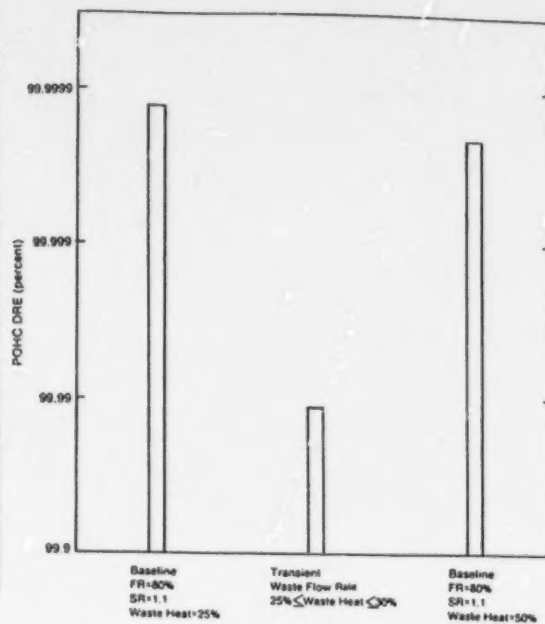


FIG. 8 DRE VERSUS TRANSIENT FLUCTUATIONS IN WASTE FLOW RATE

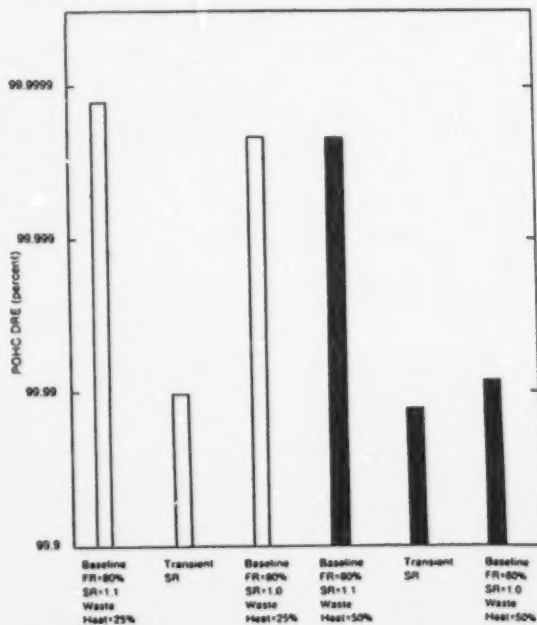


FIG. 7 DRE VERSUS TRANSIENT FLUCTUATIONS IN STOICHIOMETRIC RATIO

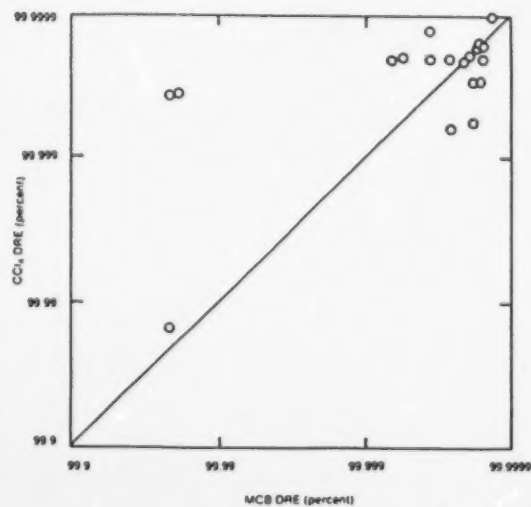


FIG. 9 CCl₄ DRE VERSUS MCB DRE

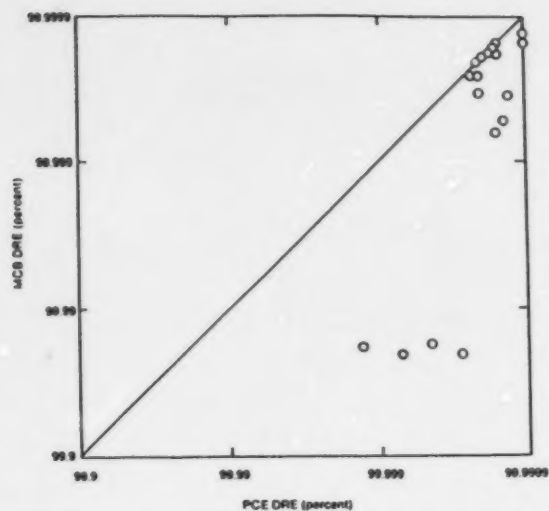


FIG. 10 MCB DRE VERSUS PCE DRE

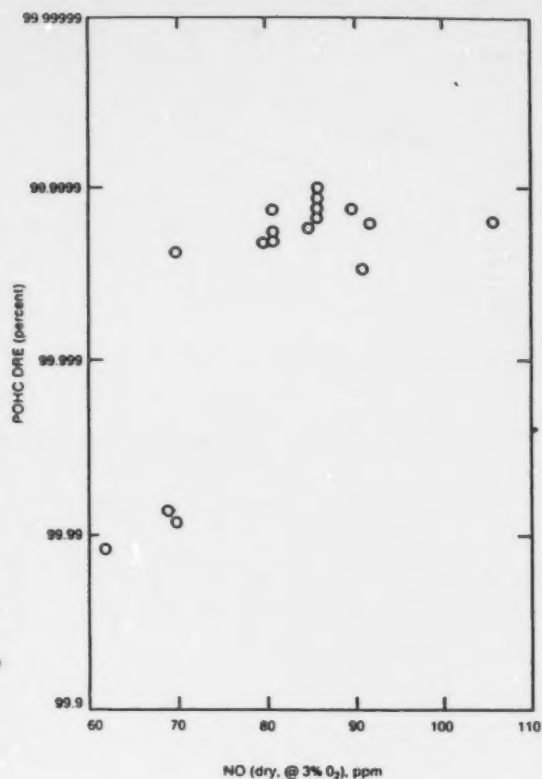


FIG. 12 POHC DRE VERSUS NO

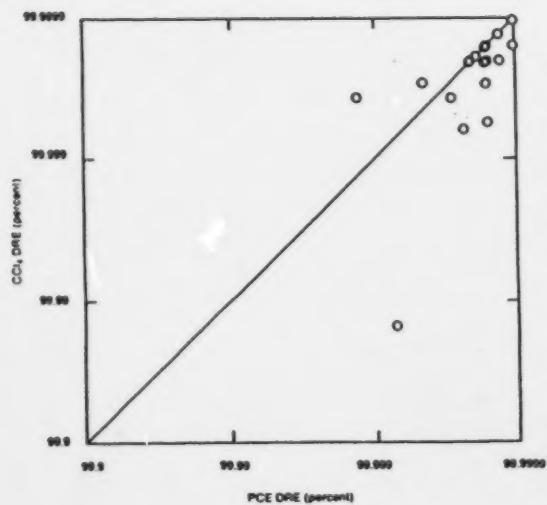


FIG. 11 CCl₄ DRE VERSUS PCE DRE

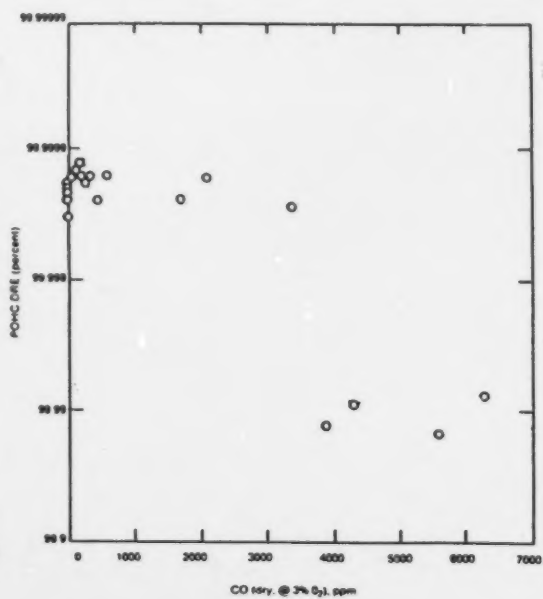


FIG. 13 POHC DRE VERSUS CO

TABLE I. PIC AND POHC EMISSIONS

Data point	CCl ₄ ($\mu\text{g}/\text{m}^3$)	MCB ($\mu\text{g}/\text{m}^3$)	PCE ($\mu\text{g}/\text{m}^3$)	POHC ($\mu\text{g}/\text{m}^3$)	CHCl ₃ ($\mu\text{g}/\text{m}^3$)	CH ₂ Cl ₂ ($\mu\text{g}/\text{m}^3$)	C ₂ HCl ₃ ($\mu\text{g}/\text{m}^3$)	C ₂ H ₄ Cl ₂ ($\mu\text{g}/\text{m}^3$)	Other PICs ($\mu\text{g}/\text{m}^3$)	PIC ($\mu\text{g}/\text{m}^3$)	PIC (μPOHC)	QPOHC ($\mu\text{m}/\text{min}$)
203.22.MV.2	2.7	<0.5	<0.5	<3.7	<0.5	-	-	-	-	-	-	0
303.21.MV.1	<0.5	<0.5	<0.5	<1.5	-	Trace	-	-	-	-	-	0.0254
303.21.MV.2	<0.5	<0.5	<0.5	<1.5	-	0.5	-	-	-	<1.0	0.67	0.0279
303.21.MV.3	<0.5	<0.5	<0.5	<1.5	-	0.8	-	-	-	<1.3	0.89	0.0279
303.21.MV.4	<0.5	<0.5	<0.5	<1.5	<0.5	2.6	-	-	-	<3.1	2.07	0.0254
303.21.MV.5	<0.5	<0.5	<0.5	<1.5	<0.5	12.6	-	-	-	<13.1	8.73	0.0263
303.17.MV1	1.2	<0.5	<0.5	<2.2	-	5.1	-	-	-	5.1	2.32	0.0254
303.22.MV1	1.4	3.6	0.7	6.6	2.4	10.2	-	-	-	12.6	2.25	0.0559
303.22.MV2	<0.5	2.6	<0.5	<3.6	<0.5	8.4	-	-	-	<8.9	2.47	0.0547
303.18.MV1	<0.5	0.6	<0.5	<1.6	<0.5	<0.5	-	-	-	<1.0	0.63	0.0517
303.18.MV2	0.9	1.1	<0.5	<2.5	<0.5	<0.5	-	-	-	<1.0	0.4	0.0517
305.37.MV2	1.3	0.7	0.6	2.6	3.9	-	-	-	-	3.9	1.5	0.0271
302.14.MV2	2.3	125	3.2	130.5	3.3	65	-	7.1	8.4	83.8	0.64	0.055
302.13.MV1	0.8	2.1	<0.5	<3.4	<0.5	0.9	-	-	-	<1.4	0.41	0.0288
302.09.MV1	0.8	<0.5	<0.5	<1.8	0.5	0.7	-	-	-	1.2	0.67	0.0263
302.09.MV2	2	0.7	<0.5	<3.2	0.8	0.9	-	-	2	3.7	1.16	0.0263
302.16.MV1	86.1	148.6	4	238.7	2.1	31.2	-	-	29	62.3	0.26	0.0616
302.16.MV1	2.6	145	2	149.6	3.5	63.8	-	-	24.6	91.9	0.61	0.0599
302.14.MV1	2.6	122	5.4	130	4	104	-	-	54.6	162.6	1.25	0.0559
302.14a.MV1	12.4	117.8	5	135.2	<1.0	147	-	-	39.2	<187.2	1.38	0.0628
402.14.MV1	20	309	110	439	6.5	90	135	174	142	547.5	1.25	0.0563
403.21.MV1	1.4	50	7.8	59.2	0.8	1.6	3.9	-	10.3	16.6	0.28	0.0282
501.4.MV1	31.4	0.5	<0.5	<32.4	Trace	-	Trace	-	-	-	-	0.0197
502.2.MV1	48.9	1.1	<0.5	<50.5	0.7	0.8	0.7	-	-	2.2	0.04	0.0209
501.2.MV1	1.8	3.9	<0.5	<6.2	0.9	1.1	-	-	1.8	3.8	0.61	0.052
502.2.MV1	0.9	3.5	<0.5	<4.9	5.6	<0.5	-	-	<0.5	<6.6	1.35	0.0507
502(R).2.MV1	10.6	2	<1.0	<13.6	2.2	1.6	-	-	-	3.8	0.28	0.0493
501(R).2.MV1	2.6	<1.0	<1.0	<4.6	1.2	2	-	-	-	3.2	0.7	0.0521
601.2.MV1	63.9	5.8	1.2	70.9	-	-	-	-	1.9	1.9	0.03	0.0286
602.2.MV1	29.8	198.2	15.2	243.2	3.1	16.8	-	7.8	35.7	63.4	0.26	0.0567
701.2.MV1	81	8.1	3.7	92.8	0.9	2.3	-	-	1	4.2	0.05	0.0296

TABLE II. SUMMARY OF POTENTIAL DRE FAILURE MODES

Load (percent)	Waste heat input (percent)	SR	Atomization	Description	POHC DRE
80	50	1.0	Good	Steady state	99.991265
80	50	1.0	Good	Steady state	99.990645
90	50	1.0	Good	Steady state	99.983056
90	50	1.0	Good	Steady state	99.989031
80	25	1.1	Poor	Steady state	99.969901
80	50	1.0	Poor	Steady state	99.967805
70 -> 90	-	1.1	Good	Ramping up of firing rate	99.983461
90 -> 70	-	1.1	Good	Ramping down of firing rate	99.990426
80	25	1.1 <-> 1.0	Good	Transient SR	99.988833
80	50	1.1 <-> 1.0	Good	Transient SR	99.980111
-	-	1.1 <-> 1.0	Good	Transient waste feedrate	99.985517
90 -> 70	-	1.1	Good	Higher ramping rate (down) of firing rate	99.996797